

# THE REVISED QUANTUM MECHANICAL THEORY OF THE OPTICAL ACTIVITY OF CRYSTALS

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**ABSTRACT:** In this paper we present the revised view on the optical activity of crystals based on the model of two damped coupled oscillators. The results are compared with the results of the same problem solved before and it is presented that the results of the quantum mechanical model are the generalization of the classical model only and they do not introduce the new terms of quantum mechanical nature. The results are discussed with regard to the rotational strengths of normal modes of vibrations and the formula for the complex rotatory power  $\bar{\rho}(\omega)$  containing the rotational strengths is presented.

## 1 Introduction

Optical activity (OA) is manifested by dissymmetric species because the transition probabilities are different for the left and the right circularly polarized wave. This means a difference between the complex refractive indices that we denote  $\bar{n}_l$  for the left and  $\bar{n}_r$  for the right circularly polarized wave. The complex rotatory power may be considered to be complex quantity given by

$$\bar{\rho} = \rho + i\sigma = \frac{\omega}{2c} (\bar{n}_l - \bar{n}_r) = \frac{\omega}{2c} [(n_l - n_r) + i(\kappa_l - \kappa_r)], \quad (1)$$

where  $\rho$  is the rotation angle of incidenting linear polarized light per unit length, and for a very excellent approximation  $\sigma$  is the ellipticity per unit length. The variation of  $\rho$  with frequency  $\omega$  or wavelength  $\lambda$  is called the optical rotatory dispersion (ORD) and the variation of  $\sigma$  is the circular dichroism (CD);  $n_l$ ,  $n_r$

and  $\kappa_l$ ,  $\kappa_r$  are real and imaginary parts of the complex refraction indices  $\bar{n}_l$  and  $\bar{n}_r$ . So that the OA has both aspects arising from interaction of radiation with matter - dispersive and absorptive and these aspects are connected by the Kramers - Kronig relations.

The important group of the optically active crystals are the crystals with screw axis of symmetry belonging to the space group of symmetry  $D_3^4$  and its enantiomorphic  $D_3^6$ . The typical representatives of these crystals are  $\alpha$ -quartz, cinnabar, tellurium, selen, camphor and benzil. The optical activity of these crystals is caused by the asymmetrical originating of the crystal structure because the molecules or atoms forming the crystals are symmetrical. Camphor is the exception because its molecules are optically active. It means that the other crystals belonging to these groups of symmetry are optically active in the crystalline state only.

The dispersion of the optical activity was studied in works based on the excitons theory [1, 2, 3, 4], on the theory of coupled oscillators [5, 6, 7, 8, 9] or on the Lagrangian formalism [10]. But all these models based on the exciton theory, some models of coupled oscillators and the model in [10] are solved in the frequency region far from the absorption range. It is well known that the CD is nozeroth only in a very narrow frequency region in the absorption range and therefore these models solve only ORD as one part of the optical activity independently on the CD.

Both aspects of optical activity can be solved by means of the model of the dumped coupled oscillators. Vyšín [8] has solved this model semiclassically and Janků [9] by quantum mechanical way where the role of dumping plays the limited lifetime of oscillators in excited states. But Janků has got some other terms in comparison with the results of [8] in his formulae for ORD and CD and he supposes that these terms are of quantum mechanical nature. At the same time the sense of these terms is not evident. For this reason we revise the solution of this problem.

All above mentioned works that solve the optical activity of crystals by the coupled oscillator model arised from the Kuhn model [11] of two coupled oscillators forming the compound oscillator. It is well known that two asymmetrically oriented harmonic oscillators coupled together form the fundamental optically active unit. With regard to the structure of crystals belonging to the space groups of symmetry  $D_3^4$  and  $D_3^6$  we can use the following Chandrasekhar model of compound oscillator.

We assume that the coordinate axis  $z$  is parallel with the crystal axis  $c$ . The first single dumped linear harmonic oscillator lies in the plane  $z = 0$  and his direction of vibrations is given by direction cosines  $\alpha$ ,  $\beta$ ,  $\gamma$ . The second oscillator lies in the plane  $z = d$  where as  $d$  we denote the projection of distance between oscillators into the  $z$  axis and his direction of vibrations is turned by the angle  $\theta$  around axis  $z$  with respect to the first one. The direction of vibrations of the second oscillator is so given by the cosines  $\alpha \cos \theta - \beta \sin \theta$ ,  $\alpha \sin \theta + \beta \cos \theta$ ,  $\gamma$ .

Both oscillators lie on the helix which is given by the structure of crystal. The angle  $\theta$  is, of course, 120 deg for the crystals belonging to the space groups of symmetry  $D_3^4$  and  $D_3^6$ .

Both oscillators forming the compound oscillator are identical. They have the identical mass, electric charge and lifetime in excited states. We take the interaction between adjacent oscillators as weak, that is for example as dipol - dipol character. We suppose that the left and the right circularly polarized wave, into which the incidenting linear polarized wave is decomposed in the optically active crystal, propagate only along the crystal axis. This is the most practically important case because the optical activity is not masked by birefringence.

## 2 The quantum mechanical solution of the model

We can set the Schrödinger equation for the one compound oscillator in the field of the left and the right circularly polarized wave

$$\hat{H}\Psi = -\frac{\hbar^2}{2m} \sum_{\xi=1}^2 \frac{\partial^2 \Psi}{\partial r_{\xi}^2} + \frac{m\omega_0^2}{2} \sum_{\xi=1}^2 r_{\xi}^2 \Psi + \mu r_1 r_2 \Psi + \frac{i}{m\omega} \sum_{\xi=1}^2 F_{\xi}^{l,r} \hat{p}_{\xi} e^{\gamma_0 t} \Psi, \quad (2)$$

where  $r_1, r_2$  are the displacements of oscillators from equilibrium,  $\mu r_1 r_2$  is the potential energy of mutual interactions of the oscillators,  $F_1^{l,r} = eE_1^{l,r}$ ,  $F_2^{l,r} = eE_2^{l,r}$  are the electric forces projections of the left and the right circularly polarized wave into the vibration directions of the oscillators,  $e$  is the charge of electron. The upper index  $l, r$  holds for the left and the right circularly polarized wave,  $\hat{p}_1$  and  $\hat{p}_2$  are the moment operators of both oscillators. The small positive parameter  $\gamma_0$  gives the possibility of the adiabatic interaction at the time  $t = -\infty$ . The dumping due to the limited lifetime of oscillators in their excited states is formally introduced by the parameter  $\gamma_0$ .

The electric vector  $\vec{E}$  of the left and the right circularly polarized wave propagating along  $z$  axis has the components

$$\begin{aligned} E_x^l &= E_0 e^{-i(\omega t - k_l z)}, & E_y^l &= E_0 e^{-i(\omega t - k_l z - \frac{\pi}{2})} \\ E_x^r &= E_0 e^{-i(\omega t - k_r z)}, & E_y^r &= E_0 e^{-i(\omega t - k_r z + \frac{\pi}{2})} \end{aligned} \quad (3)$$

and the vector  $\vec{E}$  projections of these waves into the directions of vibrations of oscillators are

$$\begin{aligned} E_1^{l,r} &= E_0 e^{-i\omega t} \left( \alpha + \beta e^{\pm i\frac{\pi}{2}} \right) \\ E_2^{l,r} &= E_0 e^{-i\omega t} \left[ (\alpha \cos \theta - \beta \sin \theta) e^{i\phi_{l,r}} + (\alpha \sin \theta + \beta \cos \theta) e^{i(\phi_{l,r} \pm \frac{\pi}{2})} \right], \end{aligned} \quad (4)$$

where  $\phi_{l,r} = k_{l,r} = \bar{n}_{l,r} \omega d / c$  is a complex phase shift and  $\bar{n}_{l,r}$  are refractive indices of medium for the left (index  $l$ ) and the right (index  $r$ ) circularly polarized wave.

In all terms in eq. (4) and further the  $+$  sign holds for the left and the  $-$  sign for the right circularly polarized wave.

Due to the mutual interaction between coupled oscillators the natural frequency  $\omega_0$  splits into two adjacent frequencies of the normal modes of vibrations. Introducing the normal coordinates  $q_1$  and  $q_2$  which are given by the relations

$$r_1 = \frac{1}{\sqrt{2}}(q_1 + q_2), \quad r_2 = \frac{1}{\sqrt{2}}(q_1 - q_2), \quad (5)$$

into eq. (2) we get

$$\begin{aligned} \hat{\mathcal{H}}\Psi = & -\frac{\hbar^2}{2m} \sum_{\eta=1}^2 \frac{\partial^2 \Psi}{\partial q_\eta^2} + \frac{m\omega_0^2}{2} \sum_{\eta=1}^2 q_\eta^2 \Psi + \frac{1}{2}\mu (q_1^2 - q_2^2) \Psi \\ & + \frac{i}{m\omega} \left[ \frac{1}{\sqrt{2}} (F_1^{l,r} + F_2^{l,r}) \hat{p}_{q_1} + \frac{1}{\sqrt{2}} (F_1^{l,r} - F_2^{l,r}) \hat{p}_{q_2} \right] e^{\gamma_0 t} \Psi. \end{aligned} \quad (6)$$

It may be easily verified that in the eq. (6) the expressions  $\frac{1}{\sqrt{2}} (F_1^{l,r} + F_2^{l,r})$  and  $\frac{1}{\sqrt{2}} (F_1^{l,r} - F_2^{l,r})$  are the projections of electric forces in normal coordinates. We denote them as  $F_{q_1}^{l,r}$  and  $F_{q_2}^{l,r}$ .

Now the equation (6) can be separated by means of

$$\begin{aligned} \Psi(q_1, q_2, t) &= \Psi_1(q_1, t) \Psi_2(q_2, t), \\ \hat{\mathcal{H}} &= \hat{\mathcal{H}}_{q_1} + \hat{\mathcal{H}}_{q_2}. \end{aligned} \quad (7)$$

Substituting (7) into (6), further dividing by  $\Psi_1(q_1, t) \Psi_2(q_2, t)$  and posing the member with  $q_1$  to  $\hat{\mathcal{H}}_{q_1}$  and  $q_2$  to  $\hat{\mathcal{H}}_{q_2}$  we get two equations

$$\begin{aligned} \hat{\mathcal{H}}_{q_1} \Psi_1(q_1, t) &= -\frac{\hbar^2}{2m} \frac{\partial^2 \Psi_1(q_1, t)}{\partial q_1^2} + \frac{m\omega_0^2 + \mu}{2} q_1^2 \Psi_1(q_1, t) \\ &\quad + \frac{i}{m\omega} (F_{q_1}^{l,r} \hat{p}_{q_1}) e^{\gamma_0 t} \Psi_1(q_1, t) \\ \hat{\mathcal{H}}_{q_2} \Psi_2(q_2, t) &= -\frac{\hbar^2}{2m} \frac{\partial^2 \Psi_2(q_2, t)}{\partial q_2^2} + \frac{m\omega_0^2 - \mu}{2} q_2^2 \Psi_2(q_2, t) \\ &\quad + \frac{i}{m\omega} (F_{q_2}^{l,r} \hat{p}_{q_2}) e^{\gamma_0 t} \Psi_2(q_2, t) \end{aligned} \quad (8)$$

and we see that the natural frequency is split into two vibrations of the normal modes. For these frequencies we have

$$\omega_1^2 = \omega_0^2 + Q, \quad \omega_2^2 = \omega_0^2 - Q; \quad (9)$$

$Q = \mu/m$ .

For  $F_{q_1}^{l,r}$  and  $F_{q_2}^{l,r}$  in the equations (8) we can derive using eq. (4) the expression

$$F_{q_\eta}^{l,r} = e(a_{q_\eta}^{l,r}) E_0 e^{-i(\omega t + \sigma_{q_\eta}^{l,r})}; \quad \eta = 1, 2. \quad (10)$$

The coefficients  $a_{q_\eta}^{l,r}$  are determined by the relations

$$\begin{aligned} \left(a_{q_1}^{l,r}\right)^2 &= \left(\alpha^2 + \beta^2\right) \left(1 + \cos \theta \mp \phi_{l,r} \sin \theta\right), \\ \left(a_{q_2}^{l,r}\right)^2 &= \left(\alpha^2 + \beta^2\right) \left(1 - \cos \theta \pm \phi_{l,r} \sin \theta\right); \end{aligned} \quad (11)$$

$\sigma_{q_\eta}^{l,r}$  are the meaning of the phase shifts only. In the eqs. (11) the upper sign holds for the left and the lower sign for the right circularly polarized wave. The eqs. (8) can be rewritten in the general form

$$\begin{aligned} \hat{\mathcal{H}}_{q_\eta} \Psi_\eta(q_\eta, t) &= -\frac{\hbar^2}{2m} \frac{\partial^2 \Psi_\eta(q_\eta, t)}{\partial q_\eta^2} + \frac{m\omega_\eta^2}{2} q_\eta^2 \Psi_\eta(q_\eta, t) \\ &+ \frac{ie}{m\omega} \left(a_{q_\eta}^{l,r}\right) E_0 \hat{p}_{q_\eta} e^{-i(\omega t + \sigma_{q_\eta}^{l,r}) + \gamma_0 t} \Psi_\eta(q_\eta, t); \end{aligned} \quad (12)$$

$\eta = 1, 2$ . The Schrödinger equations for the normal modes of vibrations then are

$$i\hbar \frac{\partial \Psi_\eta(q_\eta, t)}{\partial t} = \hat{\mathcal{H}}_{q_\eta}^{l,r^0} \Psi_\eta(q_\eta, t) + \hat{\mathcal{H}}_{q_\eta}^{l,r^p} \Psi_\eta(q_\eta, t), \quad (13)$$

where  $\hat{\mathcal{H}}_{q_\eta}^{l,r^0}$  is a nonperturbed and  $\hat{\mathcal{H}}_{q_\eta}^{l,r^p}$  a perturbed hamiltonian. In our case

$$\hat{\mathcal{H}}_{q_\eta}^{l,r^0} = -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial q_\eta^2} + \frac{m\omega_\eta^2}{2} q_\eta^2 \quad (14)$$

and we see that the nonperturbed hamiltonian depends only on the index  $\eta$  of the normal mode of vibrations and it doesn't depend on the polarization of the light wave. We can further write  $\hat{\mathcal{H}}_{q_\eta}^{l,r^0} = \hat{\mathcal{H}}_{q_\eta}^0$ . On the other hand the perturbed hamiltonian

$$\hat{\mathcal{H}}_{q_\eta}^{l,r^p} = \frac{ie}{m\omega} \left(a_{q_\eta}^{l,r}\right) E_0 \hat{p}_{q_\eta} e^{-i(\omega t + \sigma_{q_\eta}^{l,r}) + \gamma_0 t} \quad (15)$$

depends on the index of the mode and also on the polarization of the wave.

The mean value of the induced electric dipole moment from the side of the left and the right circularly polarized wave that we hold as small perturbation we can solve by means of the Kubo theorem [12]

$$\left\langle \overline{e \left(a_{q_\eta}^{l,r}\right) q_\eta} \right\rangle = \left\langle e \left(a_{q_\eta}^{l,r}\right) q_\eta \right\rangle_{\eta_0} + \frac{ie^2}{m\omega} \left(a_{q_\eta}^{l,r}\right)^2 E_0 \left\langle \left\langle \hat{q}_\eta, \hat{p}_{q_\eta} \right\rangle \right\rangle_\omega e^{-i(\omega t + \sigma_{q_\eta}^{l,r}) + \gamma_0 t}. \quad (16)$$

The first term on the right side of eq. (16) is the constant dipole moment of system. This term has no meaning for us. The expression  $\left\langle \left\langle \hat{q}_\eta, \hat{p}_{q_\eta} \right\rangle \right\rangle_\omega$  is the Fourier transform of the retarded Green function  $\left\langle \left\langle \hat{q}_\eta, \hat{p}_{q_\eta} \right\rangle \right\rangle_t$  of operators  $\hat{q}_\eta$  and  $\hat{p}_{q_\eta}$ , that is

$$\left\langle \left\langle \hat{q}_\eta, \hat{p}_{q_\eta} \right\rangle \right\rangle_\omega = \frac{1}{\hbar} \int_{-\infty}^{\infty} e^{i\omega t - \gamma_0 t} \left\langle \left\langle \hat{q}_\eta, \hat{p}_{q_\eta} \right\rangle \right\rangle_t dt. \quad (17)$$

For the retarded Green function  $\langle\langle\hat{q}_\eta, \hat{p}_{q_\eta}\rangle\rangle_t$  in the ground states of quantum system  $|\eta_0\rangle$  we hold the relation

$$\langle\langle\hat{q}_\eta, \hat{p}_{q_\eta}\rangle\rangle_t = -i\vartheta(t)\langle\eta_0|\left[\hat{\tilde{q}}_\eta(t), \hat{p}_{q_\eta}\right]|\eta_0\rangle. \quad (18)$$

In the eq. (18)  $\vartheta(t)$  is the unit step function,  $\hat{\tilde{q}}_\eta(t)$  is the operator of  $q_\eta$  in the interactions representation

$$\hat{\tilde{q}}_\eta(t) = e^{i\hat{\mathcal{H}}_{q_\eta}^0 t/\hbar} \hat{q}_\eta e^{-i\hat{\mathcal{H}}_{q_\eta}^0 t/\hbar} \quad (19)$$

and for the application of the operator in the exponent on the wave function holds

$$e^{\pm i\hat{\mathcal{H}}_{q_\eta}^0 t/\hbar}|\eta_n\rangle = e^{\pm iE_{\eta_n} t/\hbar}|\eta_n\rangle = e^{\pm i\omega_{\eta_n} t}|\eta_n\rangle. \quad (20)$$

In following rearrangement we use that for matrix elements of the production of operators  $\hat{F}_{q_\eta}$  and  $\hat{K}_{q_\eta}$  holds in the normal modes the relation

$$\langle\eta_m|\hat{F}_{q_\eta}\hat{K}_{q_\eta}|\eta_n\rangle = \sum_k \langle\eta_m|\hat{F}_{q_\eta}|\eta_k\rangle \langle\eta_k|\hat{K}_{q_\eta}|\eta_n\rangle \quad (21)$$

and further we use that the matrix elements of operator  $\hat{p}_{q_\eta}$  are

$$\langle\eta_m|\hat{p}_{q_\eta}|\eta_n\rangle = im\omega_{\eta_{mn}}\langle\eta_m|\hat{q}_\eta|\eta_n\rangle; \quad (22)$$

$$\omega_{\eta_{mn}} = \omega_{\eta_m} - \omega_{\eta_n}.$$

Then we can solve that

$$\langle\eta_0|\left[\hat{\tilde{q}}_\eta(t), \hat{p}_{q_\eta}\right]|\eta_0\rangle = im \sum_k \omega_{\eta_{k0}} |\langle\eta_k|\hat{q}_\eta|\eta_0\rangle|^2 \cdot \left(e^{-i\omega_{\eta_{k0}} t} + e^{i\omega_{\eta_{k0}} t}\right) \quad (23)$$

and after substituting the result of eq. (23) into (18) and solving the integral on the right side of eq. (17) we have

$$\langle\langle\hat{q}_\eta, \hat{p}_{q_\eta}\rangle\rangle_\omega = \frac{2i\omega m}{\hbar} \sum_k \frac{\omega_{\eta_{k0}} |\langle\eta_k|\hat{q}_\eta|\eta_0\rangle|^2}{\omega^2 - \omega_{\eta_{k0}}^2 + 2i\gamma_0\omega}. \quad (24)$$

The induced dipole moments  $d_{q_\eta}^{l,r}$  we can solve by means of (24) and (16)

$$\begin{aligned} d_{q_\eta}^{l,r} &= \sum_k \frac{2e^2 \left(d_{q_\eta}^{l,r}\right)^2 \omega_{\eta_{k0}} |\langle\eta_k|\hat{q}_\eta|\eta_0\rangle|^2}{\hbar \left(\omega_{\eta_{k0}}^2 - \omega^2 - 2i\gamma_0\omega\right)} \\ &\times E_0 e^{-i(\omega t + \sigma_{q_\eta}^{l,r}) + \gamma_0 t} \end{aligned} \quad (25)$$

and we can introduce the oscillator strengths of the normal modes of vibrations into (25) by the relation

$$f_{q_\eta} = \frac{2m\omega_{\eta_{k0}} |\langle\eta_k|q_\eta|\eta_0\rangle|^2}{\hbar} \quad (26)$$

Now the induced dipole moments can be expressed as

$$d_{q_\eta}^{l,r} = \sum_k \left(a_{q_\eta}^{l,r}\right)^2 \frac{e^2 f_{q_\eta}}{m} \frac{E_0 e^{-i(\omega t + \sigma_{q_\eta}^{l,r}) + \gamma_0 t}}{\omega_{\eta k_0}^2 - \omega^2 - 2i\gamma_0 \omega}. \quad (27)$$

The mean polarizability per unit volume would be then

$$\chi_{q_\eta}^{l,r} = \frac{N d_{q_\eta}^{l,r}}{E_0 e^{-i(\omega t + \sigma_{q_\eta}^{l,r}) + \gamma_0 t}}, \quad (28)$$

where  $N$  is the number of coupled oscillators in a volume unit. But if we include all coupling in the crystal then we see that the number of coupling between two adjacent oscillators in the direction of propagating light is the same as the number of single oscillators.

Substituting from (28) and (27) into the Drude - Sellmaier dispersion relation of refractive indices of the crystals

$$\bar{n}_{l,r}^2 - 1 = 4\pi \sum_{\eta=1}^2 \chi_{q_\eta}^{l,r} \quad (29)$$

we have

$$\bar{n}_{l,r}^2 - 1 = \frac{4\pi N e^2}{m} \sum_{\eta=1}^2 \sum_k \left(a_{q_\eta}^{l,r}\right)^2 \frac{f_{q_\eta}}{\omega_{\eta k_0}^2 - \omega^2 - 2i\gamma_0 \omega} \quad (30)$$

and we are able to solve the relation

$$\bar{n}_l^2 - \bar{n}_r^2 = \frac{4\pi N e^2}{m} \sum_{\eta=1}^2 \sum_k \frac{\left[\left(a_{q_\eta}^l\right)^2 - \left(a_{q_\eta}^r\right)^2\right] f_{q_\eta}}{\omega_{\eta k_0}^2 - \omega^2 - 2i\gamma_0 \omega} \quad (31)$$

that after substituting from (11) gives

$$\begin{aligned} \bar{n}_l^2 - \bar{n}_r^2 &= \frac{4\pi N e^2}{m} (\alpha^2 + \beta^2) \sin \theta (\phi_l + \phi_r) \\ &\times \sum_k \left[ \frac{-f_{q_1}}{\omega_{1k_0}^2 - \omega^2 - 2i\gamma_0 \omega} + \frac{f_{q_2}}{\omega_{2k_0}^2 - \omega^2 - 2i\gamma_0 \omega} \right]. \end{aligned} \quad (32)$$

We know that for the complex rotatory power we have the relation  $\bar{\rho}(\omega) = \frac{\omega}{2c} (\bar{n}_l - \bar{n}_r)$ , further  $\phi_l + \phi_r = \omega d (\bar{n}_l + \bar{n}_r) / c$  and  $\bar{n}_l^2 - \bar{n}_r^2 = (\bar{n}_l + \bar{n}_r) (\bar{n}_l - \bar{n}_r)$ . Using these relation we get for  $\bar{\rho}(\omega)$  the formula

$$\begin{aligned} \bar{\rho}(\omega) &= \frac{2\pi N d e^2}{m c^2} \omega^2 (\alpha^2 + \beta^2) \sin \theta \\ &\times \sum_k \left[ \frac{-f_{q_1}}{\omega_{1k_0}^2 - \omega^2 - 2i\gamma_0 \omega} + \frac{f_{q_2}}{\omega_{2k_0}^2 - \omega^2 - 2i\gamma_0 \omega} \right]. \end{aligned} \quad (33)$$

Now we may find the real and the imaginary part of  $\bar{\rho}(\omega)$  that from (1) are ORD and CD

$$\text{Re}\bar{\rho}(\omega) = \rho(\omega) = A\omega^2 \cdot \sum_k \left[ \frac{-f_{q1} (\omega_{1k0}^2 - \omega^2)}{(\omega_{1k0}^2 - \omega^2)^2 + 4\gamma_0^2\omega^2} + \frac{f_{q2} (\omega_{2k0}^2 - \omega^2)}{(\omega_{2k0}^2 - \omega^2)^2 + 4\gamma_0^2\omega^2} \right], \quad (34)$$

$$\text{Im}\bar{\rho}(\omega) = \sigma(\omega) = 2A\gamma_0\omega^3 \cdot \sum_k \left[ \frac{-f_{q1}}{(\omega_{1k0}^2 - \omega^2)^2 + 4\gamma_0^2\omega^2} + \frac{f_{q2}}{(\omega_{2k0}^2 - \omega^2)^2 + 4\gamma_0^2\omega^2} \right], \quad (35)$$

where  $A = \frac{2\pi Nde^2}{mc^2}(\alpha^2 + \beta^2) \sin \theta$ .

We know that the quantum transition is optically active when it has a nonzero value of the rotational strength. It is well known that the formulae for ORD and CD are often expressed as the sum of products of rotational strengths for different transitions and frequency dependent factors [13]. Our formulae (34) and (35) for ORD and CD do not contain the rotational strengths explicitly. But we have showed in the paper [14] that we can express our formulae in terms of rotational strengths in a similar way.

For a transition from some ground state  $|0\rangle$  to the excited state  $|k\rangle$  the rotational strength is given by imaginary part of the scalar products of electric and magnetic dipole moments  $\langle 0|\hat{\vec{d}}|k\rangle$  and  $\langle k|\hat{\vec{m}}|0\rangle$ . The rotational strength is so

$$R_{0k} = \text{Im} \left( \langle 0|\hat{\vec{d}}|k\rangle \langle k|\hat{\vec{m}}|0\rangle \right). \quad (36)$$

In our case of coupled oscillators the ground and the excited state are split into two states  $|\eta_0\rangle$  and  $|\eta_k\rangle$ ,  $\eta = 1, 2$ . Therefore the rotational strengths must be considered between corresponding ground and excited states and they are

$$R_{\eta_0k} = \text{Im} \left( \langle \eta_0|\hat{\vec{d}}_{q_\eta}|\eta_k\rangle \langle \eta_k|\hat{\vec{m}}_{q_\eta}|\eta_0\rangle \right); \quad \eta = 1, 2 \quad (37)$$

and for these rotational strengths we have derived in [14] the results

$$R_{10k} = -\frac{\hbar e^2 d(\alpha^2 + \beta^2) f_{q1} \sin \theta}{8mc} \quad (38)$$

and

$$R_{20k} = \frac{\hbar e^2 d(\alpha^2 + \beta^2) f_{q2} \sin \theta}{8mc}. \quad (39)$$

We see that all terms of those rotational strengths that depends on the crystal structure are comprehended in the formula for the complex rotatory power (33) and consequentially in the formulae (34) and (35) for ORD and CD.



### 3 Discussion

In the relations (34) and (35) we restrict only to one optically active quantum mechanical transition from the ground states  $|\eta_0\rangle$  to the first excited states  $|\eta_1\rangle$ . In this case we have for the transition frequencies the relations  $\omega_{110}^2 = \omega_1^2 = \omega_0^2 + Q$  and  $\omega_{210}^2 = \omega_2^2 = \omega_0^2 - Q$ .

The formulae (34) and (35) can be rewritten in the following way. The results for two-oscillator model of optical activity are often expressed as the sum of two terms. The first term contains the difference of oscillators strengths  $f_{q_2} - f_{q_1}$  and the second one the sum  $f_{q_1} + f_{q_2}$ . If we neglect the members containing  $Q^2$  and  $Q\gamma_0^2$  we can write for ORD and CD the relations

$$\rho(\omega) = A\omega^2 \left\{ \frac{(f_{q_2} - f_{q_1})(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + 4\gamma_0^2\omega^2} + \frac{Q(f_{q_1} + f_{q_2})[(\omega_0^2 - \omega^2)^2 - 4\gamma_0^2\omega^2]}{[(\omega_0^2 - \omega^2)^2 + 4\gamma_0^2\omega^2]^2} \right\}, \quad (40)$$

$$\sigma(\omega) = 2A\gamma_0\omega^2 \left\{ \frac{f_{q_2} - f_{q_1}}{(\omega_0^2 - \omega^2)^2 + 4\gamma_0^2\omega^2} + \frac{2Q(f_{q_1} + f_{q_2})(\omega_0^2 - \omega^2)}{[(\omega_0^2 - \omega^2)^2 + 4\gamma_0^2\omega^2]^2} \right\}, \quad (41)$$

but these results are formally the same as the results for the classical model of the optical activity based on the model of two coupled oscillators [8]. We assume that the terms, which Janku has obtained in the quantum mechanical model [9] in addition to the classical model, are only the consequence of used solution method.

The results of the solution of the rotational strengths (38) and (39) enable us to express the results for  $\bar{\rho}(\omega)$ ,  $\rho(\omega)$  and  $\sigma(\omega)$ , they are given by formulae (33), (34) and (35), in another form. The formula for the complex rotatory power can be expressed as

$$\bar{\rho}(\omega) = \frac{16\pi N\omega^2}{\hbar c} \sum_{\eta=1}^2 \sum_k \frac{R_{\eta_{0k}}}{\omega_{\eta_{k0}}^2 - \omega^2 - 2i\gamma_0\omega} \quad (42)$$

and similarly the formulae for  $\rho(\omega)$  and  $\sigma(\omega)$  are

$$\rho(\omega) = \frac{16\pi N\omega^2}{\hbar c} \sum_{\eta=1}^2 \sum_k \frac{(\omega_{\eta_{k0}}^2 - \omega^2) R_{\eta_{0k}}}{(\omega_{\eta_{k0}}^2 - \omega^2)^2 + 4\gamma_0^2\omega^2}, \quad (43)$$

$$\sigma(\omega) = \frac{32\pi N\omega^3\gamma_0}{\hbar c} \sum_{\eta=1}^2 \sum_k \frac{R_{\eta_{0k}}}{(\omega_{\eta_{k0}}^2 - \omega^2)^2 + 4\gamma_0^2\omega^2}. \quad (44)$$

We can accept these results as the crystal analogs of the similar results which we know from the optical activity of molecules [15] or recently of delocalized molecular aggregates [16]. In the future it will be verified if these results are

valid also for other models of coupled oscillators in the crystal optical activity or by what way they must be modified.

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